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(71) Applicant: **The British Petroleum Company p.l.c.**
Britannic House Moor Lane
London EC2Y 9BU (GB)

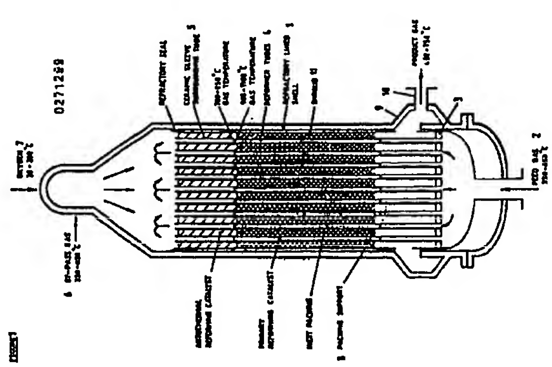
(72) Inventor: **Hardmann, Stephen British Petroleum Company p.l.c.**
Britannic House West Moor Lane
London EC2Y 9BU (GB)

McFarlane, Roderick Arthur
19 High View
Cheam Surrey SM2 7DZ (GB)

(74) Representative: **Richardson, Derek et al**
BP INTERNATIONAL LIMITED Patents & Agreements Division Chertsey Road
Sunbury-on-Thames Middlesex TW16 7LN (GB)

(54) Apparatus and use thereof in the production of synthesis gas.

(57) An apparatus for the production of synthesis gas from a hydrocarbon feed gas which apparatus comprises an outer shell (1), a first inlet (2) at the bottom of the shell for the introduction of preheated gaseous hydrocarbon feed, steam and optionally carbon dioxide, a steam reforming zone comprising a plurality of tubes (4) together forming a bundle extending upwardly from a tubesheet (3) communicating with the first inlet (2), the tubes on their tubeside defining three portions (i) a lower empty portion, (ii) an intermediate portion adapted to contain steam reforming catalyst to effect a first steam reforming reaction, and (iii) an upper empty portion, a combustion zone comprising an oxygen mixer or mixer/burner located in the topmost region of the shell communicating with a second inlet (7) for combustion of hydrocarbon fed therethrough to produce hot combustion gases and thereby heat the first steam reforming reaction product, an autothermal reforming zone defined by the shellside of the upper empty portion (iii) of the steam reforming tubes and the shell adapted to contain autothermal reforming catalyst to effect a second reforming of the first steam reforming reaction product and combustion product and thereby produce heat for heating by exchange the first steam reforming reaction product in the portion (ii) of the steam reforming tubes, a heat exchange zone defined by the shellside of the intermediate portion (ii) of the steam reforming tubes and the shell adapted to contain an inert solid material for supporting the autothermal reforming catalyst, for supporting the steam reforming tube bundle and for assisting heat transfer between the shellside and the tubeside thereby heating gases on the tubeside and cooling synthesis gas from the autothermal reforming zone, and an outlet (10) for synthesis gas exiting from the heat exchange zone.



Description

APPARATUS AND USE THEREOF IN THE PRODUCTION OF SYNTHESIS GAS

The present invention relates to an apparatus for the production of synthesis gas, and to its use in the production of synthesis gas from a gaseous hydrocarbon feed.

The term synthesis gas is generally applied to a gaseous mixture principally comprising carbon monoxide and hydrogen, but also possibly containing carbon dioxide and minor amounts of methane and nitrogen, though the term is often also applied to gaseous mixtures principally comprising nitrogen and hydrogen, useful in the production of ammonia. It is with the former meaning of the term that the present invention is principally but not exclusively concerned. Synthesis gas is used, or is potentially useful, as feedstock in a variety of large-scale chemical processes, for example the production of methanol and the production of gasoline boiling range hydrocarbons by the Fischer-Tropsch process.

The production of synthesis gas is generally accomplished by either steam reforming, which includes convective reforming, autothermal reforming or partial oxidation of light hydrocarbons. Steam reforming is a highly endothermic reaction which is generally carried out in multitubular reactors with excess steam to prevent carbon build-up on the catalyst. Steam reforming generally produces synthesis gas having a hydrogen to carbon monoxide ratio of at least 3:1 depending upon the excess steam used, which ratio being higher than that required for subsequent conversion into, for example methanol (ratio of 2:1), requires further adjustment for this purpose. Non-catalytic partial oxidation is effected in burners specially designed for partial combustion of rich fuel/oxygen mixtures. In order to achieve useful conversions and product gas compositions, however, it is necessary to use more than the stoichiometric amount of oxygen, which results in higher exit gas temperatures and associated expense in oxygen plant and downstream cooling to temperatures suitable for conversion. Autothermal reforming may be regarded as a combination of steam-reforming and partial oxidation wherein the heat of reaction for highly endothermic reforming reactions is supplied in part by partial combustion of the feed in oxygen. Excess steam is also necessary in this process to limit coke deposition on the catalyst.

It has already been proposed to combine certain of these reactions within a single reaction vessel, see for example EP-A-0124226 and GB-A-2153382, with associated advantage in internal heat recovery or utilisation.

EP-A-0124226 discloses apparatus for the endothermic reaction of a hydrocarbon or hydrocarbon derivative with steam and/or carbon dioxide comprising:

an outer shell;

mutally parallel first and second tube plates disposed across the shell and dividing it into three successive zones, namely a heat exchange zone, a reactants feed zone and a products offtake zone;

at least one relatively wide tube extending into the heat exchange zone from the first tube plate bounding that zone and closed at its extremity within the zone;

at least one relatively narrow tube extending from the second tube plate into the extremity of each relatively wide tube; and

a steam reforming catalyst in the annular space between the tube.

The primary application of this apparatus is the production of synthesis gas for ammonia synthesis. In this apparatus, which includes autothermal and convective reforming in the same vessel with packing on the shellside of the reformer tubes, the partially reformed gas exiting the steam reforming catalyst is cooled via an internal concentric return tube and leaves via a second tubesheet before being transferred <u>externally</u> to the autothermal reforming section at the top of the vessel.

GB-A-2153382 discloses an autothermal reactor for the production of a synthesis gas, comprising a heat exchange chamber, a first inlet for the introduction of steam and feed gas to said heat exchange chamber, reaction tubes mounted within said heat exchanger chamber and adapted to contain catalyst therein to effect a first reforming reaction, means communicating with said reaction tubes to pass the thus reacted gases from said tubes to a combustion reaction chamber, a second inlet to introduce oxygen or oxygen-enriched air to said combustion reaction chamber, a partition separating said heat exchange chamber from said combustion chamber, said partition including means to permit the passage of combustion reaction effluent therethrough and adapted to provide a second catalyst zone, whereby the combustion reaction effluent can pass through said partition and said second catalyst zone to undergo additional reforming reaction and to produce said synthesis gas, an outlet for removal of said synthesis gas positioned in said reactor approximately adjacent said first inlet, to permit said synthesis gas to pass about the outside of said reaction tubes prior to removal through said outlet and provide heat for said first reforming reaction within said reaction tubes and cooling of said synthesis gas. The apparatus combines convective and autothermal reforming within a single shell. The primary application of the apparatus is the production of synthesis gas for ammonia production. In this apparatus shell side heat transfer is assisted by baffles. It is anticipated that the presence of these baffles is likely to cause problems in the accomodation of thermal expansion. The combustion part of the autothermal reformer is unusual in that it is at the bottom of the vessel underneath a refractory arch. This arch will be subjected to very high temperatures and could be prone to collapse.

We have now developed an apparatus for the production of synthesis gas in which convective steam reforming and autothermal reforming are

combined into a single shell in a manner which eliminates external transfer piping and substantially overcomes the potential problems associated with the apparatus of GB-A-2153382.

Accordingly, the present invention provides an apparatus for the production of synthesis gas from a hydrocarbon feed gas which apparatus comprises

an outer shell,

a first inlet at the bottom of the shell for the introduction of preheated gaseous hydrocarbon feed, steam and optionally carbon dioxide,

a steam reforming zone comprising a plurality of tubes together forming a bundle extending upwardly from a tubesheet communicating with the first inlet, the tubes on their tubeside defining three portions (i) a lower empty portion, (ii) an intermediate portion adapted to contain steam reforming catalyst to effect a first steam reforming reaction, and (iii) an upper empty portion,

a combustion zone comprising an oxygen mixer or mixer/burner located in the topmost region of the shell communicating with a second inlet for combustion of hydrocarbon fed therethrough to produce hot combustion gases and thereby heat the first steam reforming reaction product,

an autothermal reforming zone defined by the shellside of the upper empty portion (iii) of the steam reforming tubes and the shell adapted to contain autothermal reforming catalyst to effect a second reforming of the first steam reforming reaction product and combustion product and thereby produce heat for heating by exchange the first steam reforming reaction product in the portion (ii) of the steam reforming tubes,

a heat exchange zone defined by the shellside of the intermediate portion (ii) of the steam reforming tubes and the shell adapted to contain an inert solid material for supporting the autothermal reforming catalyst, for supporting the steam reforming tube bundle and for assisting heat transfer between the shellside and the tubeside thereby heating gases on the tubeside and cooling synthesis gas from the autothermal reforming zone, and

an outlet for synthesis gas exiting from the heat exchange zone.

The apparatus in use will contain steam reforming catalyst, autothermal reforming catalyst and inert solid material.

Accordingly, in another embodiment the invention provides an apparatus for the production of synthesis gas from a hydrocarabon feed gas which apparatus comprises:

an outer shell,

a first inlet at the bottom of the shell for the introduction of preheated gaseous hydrocarbon feed, steam and optionally carbon dioxide,

a steam reforming zone comprising a plurality of tubes together forming a bundle extending upwardly from a tubesheet communicating with the first inlet, the tubes on their tubeside defining three portions (i) a lower empty portion, (ii) an intermediate portion containing steam reforming catalyst to effect a first reforming reaction, and (iii) an open ended upper empty portion,

a second inlet at the top of the shell for the introduction of hydrocarbon, and optionally steam and/or carbon dioxide,

a combustion zone comprising an oxygen mixer or mixer/burner located in the topmost region of the shell communicating with the second inlet for combustion of hydrocarbon fed therethrough to produce hot combustion gases and thereby heat the first steam reforming reaction product,

an autothermal reforming zone defined by the shellside of the upper empty portion (iii) of the steam reforming tubes and the shell containing autothermal reforming catalyst to effect a second reforming of the first steam reforming reaction product and combustion product and thereby produce heat for heating by exchange the first steam reforming reaction product in the portion (ii) of the steam reforming tubes.

a heat exchange zone defined by the shellside of the intermediate portion (ii) of the steam reforming tubes and the shell containing an inert solid material for supporting the steam reforming tube bundle and for assisting heat transfer between the shellside and the tubeside of the tubes thereby heating gases on the tubeside and cooling synthesis gas from the autothermal reforming zone,

an outlet for synthesis gas exiting from the heat exchange zone.

The aforesaid apparatus affords a number of advantages as follows:

(i) By combining convective steam reforming and autothermal reforming into a single shell in a manner which eliminates external transfer piping, costs and heat losses can be reduced,

(ii) heat transfer is improved by the presence of packing on the shellside as well as on the tubeside of the tube bundle,

(iii) true countercurrent heat transfer is achievable,

(iv) a low pressure differential is achievable across the tube walls, particularly at the hot end of the tube bundle, and

(v) the inert solid material holds the tubes in position radially without significant restraining axial movement due to thermal expansion.

The outer shell may suitably be lined internally with a refractory material and may be further protected by the provision of an external cooling jacket.

Any steam reforming catalyst may be employed, suitably in a conventional ring form or in other form adapted to minimise compacting thereof.

The oxygen mixer/burner may be of conventional design and can be obtained commercially from a variety of sources. Oxygen may be fed to the mixer burner through the second inlet or through a separate inlet. Combustion may be assisted by the provision of a layer of ignition catalyst on top of the autothermal reforming catalyst.

Any autothermal reforming catalyst may be employed. The portion (iii) of the steam reforming tubes extending through the autothermal reforming zone into th combustion zone may suitably be protected from the elevated t mperatur s encountered therein by the provision of external ceramic sleeves, which may also be adapted at the top thereof to assist gas mixing in the combustion zon and at the bottom

thereof to assist the location of the tubes.

A portion of the inert solid material of the heat exchange zone may be replaced by a carbon monoxide shift conversion catalyst, in order to release the exothermic heat of this r action and thereby increase the temperature of the reactant gases in the steam reforming tube bundle. The inert solid material is preferably supported at a point above the tubesheet, preferably at a level corresponding approximately to the interface between the portion (i) and the portion (ii) of the tubes.

The steam reforming tube bundle and packing may in a preferred embodiment be housed within a cylindrical shroud spaced from the refractory lining of the outer shell by a small annular gap. The gap is preferably sealed at the top end thereof with refractory material. The shroud can serve two purposes. Firstly, it ensures that the radial pressure from the cyclic expansion and contraction of the tubes is taken on an unpressurised internal component and not the pressure shell and its refractory lining. This has the advantage that it makes the apparatus inherently much safer. Secondly, with the shroud fitted, the bundle and the catalyst can be extracted from the pressure shell as one. Once the shroud has been either cut or pulled off, the catalyst can be removed reasonably easily from the tubes because the bundle is exposed along its full length. There are large diameter holes in the shroud through which the gas leaves the bundle.

The outlet for synthesis gas exiting from the heat exchange zone may suitably be provided in an annular collector adapted to collect the synthesis gas, which collector may suitably be located at a level corresponding to the portion (i) of the tubes.

In another embodiment the invention provides a process for the production of synthesis gas from a gaseous hydrocarbon feed utilising the apparatus as hereinbefore described which process comprises:

feeding preheated gaseous hydrocarbon, steam and optionally carbon dioxide to the apparatus via the first inlet,

in the portion (ii) of the tube bundle of the steam reforming zone effecting a first reforming reaction at elevated temperature to produce a first reformed gaseous mixture comprising hydrogen, carbon monoxide, carbon dioxide, water and unreacted hydrocarbon, the gaseous mixture passing through the portion (iii) of the tube bundle into the combustion zone wherein it mixes with hot combustion gases produced by combustion of further gaseous hydrocarbon, and optionally steam and carbon dioxide, fed to the reactor through the second inlet, thereby heating the first reformed gaseous mixture,

the heated first reformed gaseous mixture and combustion gases passing to the autothermal reforming zone wherein they undergo a second reforming reaction to produce a second reformed gaseous mixture principally comprising hydrogen, carbon monoxide, carbon dioxide and water, the exothermic heat of reaction utilised to heat by exchange the first reformed gaseous mixture in the portion (ii) of the tube bundle,

the second reformed gaseous mixture passing to the heat exchange zone wherein it imparts heat by exchange to the gaseous reactant in the portion (ii) of the tube bundle and is thereby cooled, and finally withdrawing the cooled second reformed gaseous mixture through the outlet.

The gaseous hydrocarbon feedstock may suitably be a gaseous paraffinic hydrocarbon, or a mixture thereof. A preferred mixture is natural gas, the composition of which may vary according to its origin, but generally principally comprises methane together with minor proportions of ethane and propane. Before entry into the apparatus the feedstock is preheated in conventional manner.

The second reformed gaseous mixture withdrawn from the outlet of the apparatus may be treated in conventional manner to obtain synthesis gas suitable for further conversion, for example into methanol or gasoline range hydrocarbons. Generally, such treatment involves further cooling by way of steam generation and optionally carbon dioxide removal. The steam and carbon dioxide (if any) obtained thereby may suitably be recycled to the process.

The invention will now be further illustrated by reference to Figures 1 and 2 of the Drawings in which:-

Figure 1 is a schematic representation of an apparatus according to the invention.

Figure 2 is a schematic representation of a preferred form of the upper portion of a steam reformer tube as shown in Figure 1 with particular emphasis on the protection thereof.

With reference to Figure 1, 1 is a refractory lined outer shell, 2 is a feed gas inlet, 3 is a tubesheet, 4 are steam reformer tubes, 5 are external ceramic sleeves, 6 is a second gas inlet, 7 is an oxygen inlet, 8 is an inert solid support, 9 is an annular collector, 10 is an outlet and 11 is a cylindrical shroud.

With reference to Figure 2, 4 is a steam reformer tube, 5 is an external ceramic sleeve, 12 is a cast refractory cap, 13 is a ceramic fibre cylinder, 14 is a metal or ceramic grid and 15 is a device for fixing the ceramic sleeve 5 to the steam reformer tube 4.

Preheated (350 to 650°C) natural gas, together with steam and optionally recycled carbon dioxide is fed to the shell 1 through the inlet 2 and thereafter to the steam reforming tube bundle via an inlet channel. The steam reforming tube bundle consists of a plurality of tubes 4 upwardly extending from a tubesheet 3. The tubes 4 are divided into three portions (i) a lower empty portion, (ii) an intermediate portion packed with steam reforming catalyst, and (iii) an upper empty portion open at the top having an external ceramic sleeve 5 and extending into and through a bed of autothermal reforming catalyst on the shellside thereof. A preferred form of the upper portion of one of the steam reformer tubes as shown in Figure 1 and its protection is illustrated in Figure 2. In this preferred form, the tube 4 is capped with a cast refractory cap 12 and between the tube 4 and the ceramic sleeve 5 there is interposed a ceramic fibre cylinder 13. The ceramic sleeve 5 is attached to the tube 4 by a fixing device 15, which preferably takes the form of a bayonet for

easy removal and attachment. An additional preferred feature is a metal or ceramic grid 14 to limit catalyst movement at the surface.

The natural gas flows upwardly through the tubes 4 wherein its temperature is raised to about 700-850°C in the portion (ii) and undergoes a first reforming reaction catalysed by the steam reforming catalyst on the tubeside thereof to produce a mixture of hydrogen, carbon monoxide, carbon dioxide, steam and methane.

Further preheated (350 to 650°C) natural gas is fed through inlet 6 and is combusted with an oxygen-containing gas (temperature ranging from ambient to about 250°C) fed through inlet 7 in an oxygen mixer/burner (not shown) to produce a hot combustion product which mixes with the first reforming reaction product, thereby raising its temperature, before the whole gaseous mixture flows downwards through a bed of autothermal reforming catalyst. In this bed a second reforming reaction is effected at a temperature of about 900 to 1110°C and a pressure in the region of 20-80 bar to produce a second reformed gaseous mixture comprising CO, CO_2, H_2, H_2O, together with some residual methane and possibly also inert gases such as nitrogen and argon.

The gases continue to flow downwards on the shellside of the tubesbundle through a bed of inert packing material retained by a support 8, the inert packing itself serving to support the autothermal reforming catalyst bed, to locate the tubes 4 and to improve the heat transfer to the tubes.

The cooled product gases (temperature in the region 450 -750°C) leaving the inert packing material are collected via the annular shroud 9 and leave the shell through the outlet 10.

The cylindrical shroud 11 housing the tube bundle and packing takes the radial pressure arising from the expansion and contraction of the tubes and facilitates extraction of the tube bundle and catalyst in a unitary manner.

Claims

1 An apparatus for the production of synthesis gas from a hydrocarbon feed gas which apparatus comprises
an outer shell,
a first inlet at the bottom of the shell for the introduction of preheated gaseous hydrocarbon feed, steam and optionally carbon dioxide,
a steam reforming zone comprising a plurality of tubes together forming a bundle extending upwardly from a tubesheet communicating with the first inlet, the tubes on their tubeside defining three portions (i) a lower empty portion, (ii) an intermediate portion adapted to contain steam reforming catalyst to effect a first steam reforming reaction, and (iii) an upper empty portion,
a combustion zone comprising an oxygen mixer or mixer/burner located in the topmost region of the shell communicating with a second inlet for combustion of hydrocarbon fed therethrough to produce hot combustion gases and thereby heat th first steam reforming reaction product,
an autothermal reforming zone defined by the shellside of the upper empty portion (iii) of the steam reforming tubes and the shell adapted to contain autothermal reforming catalyst to effect a second reforming of the first steam reforming reaction product and combustion product and thereby produce heat for heating by exchange the first steam reforming reaction product in the portion (ii) of the steam reforming tubes,
a heat exchange zone defined by the shellside of the intermediate portion (ii) of the steam reforming tubes and the shell adapted to contain an inert solid material for supporting the autothermal reforming catalyst, for supporting the steam reforming tube bundle and for assisting heat transfer between the shellside and the tubeside thereby heating gases on the tubeside and cooling synthesis gas from the autothermal reforming zone, and
an outlet for synthesis gas exiting from the heat exchange zone.

2 An apparatus for the production of synthesis gas from a hydrocarbon feed gas which apparatus comprises:
an outer shell,
a first inlet at the bottom of the shell for the introduction of preheated gaseous hydrocarbon feed, steam and optionally carbon dioxide,
a steam reforming zone comprising a plurality of tubes together forming a bundle extending upwardly from a tubesheet communicating with the first inlet, the tubes on their tubeside defining three portions (i) a lower empty portion, (ii) an intermediate portion containing steam reforming catalyst to effect a first reforming reaction, and (iii) an open ended upper empty portion,
a second inlet at the top of the shell for the introduction of hydrocarbon, and optionally steam and/or carbon dioxide,
a combustion zone comprising an oxygen mixer or mixer/burner located in the topmost region of the shell communicating with the second inlet for combustion of hydrocarbon fed therethrough to produce hot combustion gases and thereby heat the first steam reforming reaction product,
an autothermal reforming zone defined by the shellside of the upper empty portion (iii) of the steam reforming tubes and the shell containing autothermal reforming catalyst to effect a second reforming of the first steam reforming reaction product and combustion product and thereby produce heat for heating by exchange the first steam reforming reaction product in the portion (ii) of the steam reforming tubes.
a heat exchange zone defined by the shellside of the intermediate portion (ii) of the steam reforming tubes and the shell containing an inert solid material for supporting the steam reforming tube bundle and for assisting heat

transfer between the shellside and the tubeside of the tubes thereby heating gases on the tubeside and cooling synthesis gas from the autothermal reforming zone,
an outlet for synthesis gas exiting from the heat exchange zone.

3 An apparatus according to either claim 1 or claim 2 wherein the outer shell is lined internally with a refractory material.

4 An apparatus according to any one of the preceding claims wherein the outer shell is provided with a cooling jacket.

5 An apparatus according to any one of claims 2 to 4 wherein combustion of hydrocarbon in the combustion zone is assisted by the provision of a layer of ignition catalyst on top of the autothermal reforming catalyst.

6 An apparatus according to any one of the preceding claims wherein the portion (iii) of the steam reforming tubes extending through the autothermal reforming zone into the combustion zone is provided with external ceramic sleeves for protection from the elevated temperatures encountered therein.

7 An apparatus according to claim 6 wherein the ceramic sleeves are adapted at the top thereof to assist gas mixing in the combustion zone.

8 An apparatus according to any one of claims 2 to 7 wherein a portion of the inert solid material of the heat exchange zone is replaced by a carbon monoxide shift conversion catalyst to release the exothermic heat of this reaction and thereby increase the temperature of the reactant gases in the steam reforming tube bundle.

9 An apparatus according to any one of claims 2 to 8 wherein the inert solid material of the heat exchange zone is supported at a point above the tubesheet at a level corresponding approximately to the interface between the portion (i) and the portion (ii) of the tubes.

10 An apparatus according to any one of claims 3 to 9 wherein the steam reforming tube bundle and packing is housed within a cylindrical shroud spaced from the refractory lining of the outer shell by a small annular gap.

11 An apparatus according to claim 10 wherein the annular gap is sealed at the top end thereof with refractory material.

12 An apparatus according to any one of the preceding claims wherein the outlet for synthesis gas exiting from the heat exchange zone is provided in an annular collector adapted to collect synthesis gas.

13 A process for the production of synthesis gas from a gaseous hydrocarbon feed utilising the apparatus as claimed in claims 2 to 12 which process comprises:
feeding preheated gaseous hydrocarbon, steam and optionally carbon dioxide to the apparatus via the first inlet,
in the portion (ii) of the tube bundle of the steam reforming zone effecting a first reforming reaction at elevated temperature to produce a first reformed gaseous mixture comprising hydrogen, carbon monoxide, carbon dioxide, water and unreacted hydrocarbon, the gaseous mixture passing through the portion (iii) of the tube bundl into the combustion zone wherein it mixes with hot combustion gases produced by combustion of further gaseous hydrocarbon, and optionally steam and carbon dioxide, fed to the reactor through the second inlet, thereby heating the first reformed gaseous mixture,
the heated first reformed gaseous mixture and combustion gases passing to the autothermal reforming zone wherein they undergo a second reforming reaction to produce a second reformed gaseous mixture principally comprising hydrogen, carbon monoxide, carbon dioxide and water, the exothermic heat of reaction utilised to heat by exchange the first reformed gaseous mixture in the portion (ii) of the tube bundle,
the second reformed gaseous mixture passing to the heat exchange zone wherein it imparts heat by exchange to the gaseous reactant in the portion (ii) of the tube bundle and is thereby cooled, and finally
withdrawing the cooled second reformed gaseous mixture through the outlet.

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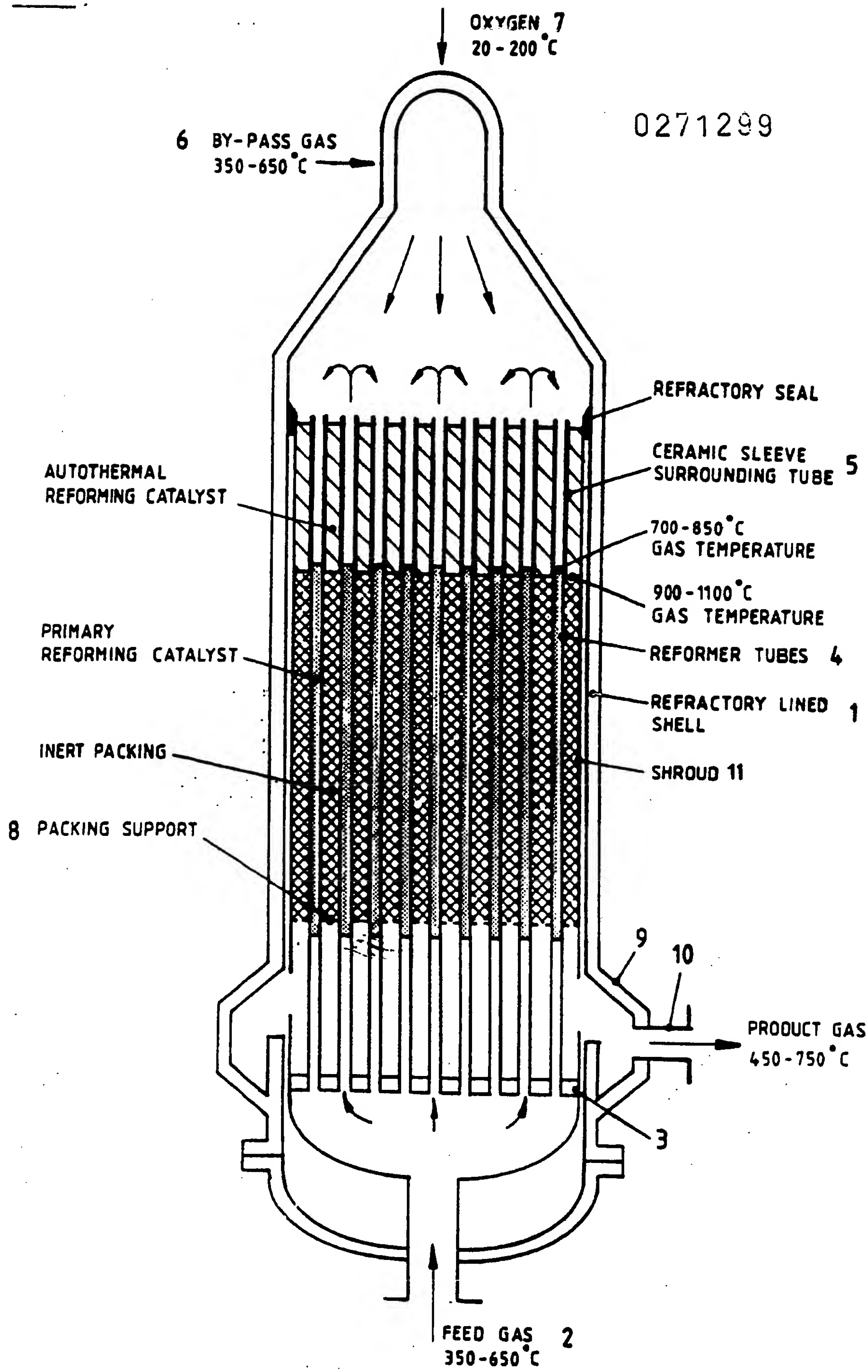
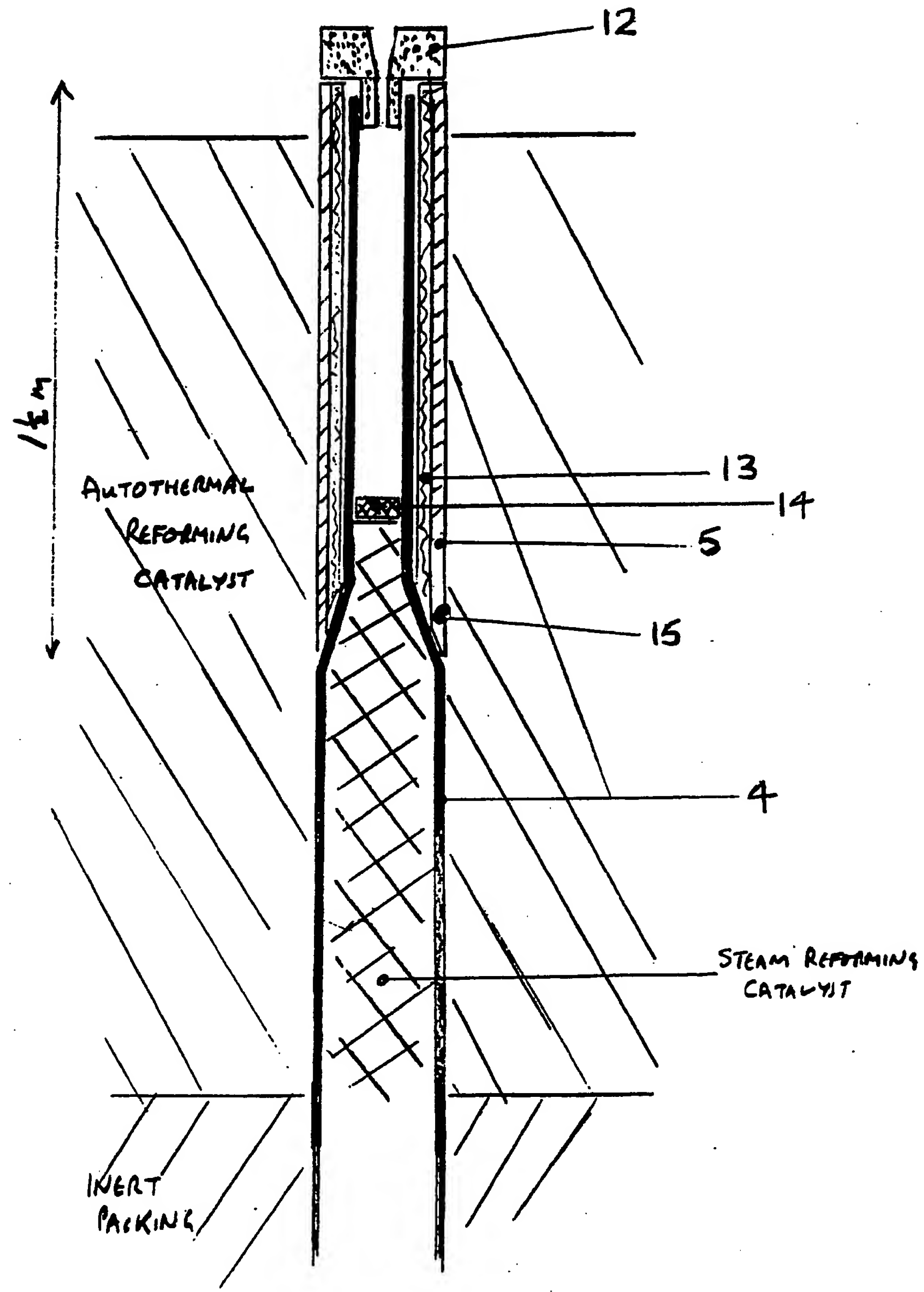


OXYGEN 7
20 - 200 °C
6 BY-PASS GAS
350-650 °C
REFRACTORY SEAL
CERAMIC SLEEVE
SURROUNDING TUBE 5
AUTOTHERMAL
REFORMING CATALYST
700-850 °C
GAS TEMPERATURE
900-1100 °C
GAS TEMPERATURE
PRIMARY
REFORMING CATALYST
REFORMER TUBES 4
REFRACTORY LINED
SHELL 1
INERT PACKING
SHROUD 11
8 PACKING SUPPORT
9
10
PRODUCT GAS
450-750 °C
3
FEED GAS 2
350-650 °C

FIGURE 2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AUTOTHERMAL REFORMING CATALYST
STEAM REFORMING CATALYST
INERT PACKING